



VERIFICATION OF TRANSLATION

Re: U.S. PATENT APPLICATION S.N.09/854,067

I, Yumi KUJIME, of Kitahama TNK Building.

7-1, Dosho-machi 1-chome, Chuo-ku.

Osaka 541-0045, Japan

hereby declare that I am the translator of the
document attached and certify that the following is
true translation to the best of my knowledge and
belief.

Signature of translator

Yumi Kujime
Yumi KUJIME

Dated this 11th day of October, 2001



ORGANIC EL DEVICE AND METHOD FOR ITS MANUFACTURE

FIELD OF THE INVENTION

The present invention relates to a novel organic EL device and a method of manufacturing the same.

BACKGROUND OF THE INVENTION

The organic EL (electroluminescent) device is a structural entity comprising an organic light-emitting layer comprised of an organic compound as sandwiched between a pair of electrodes. The organic EL device has various characteristics such as high-luminance emission, high-speed response, high energy-converting rate, and high color development and, as such, is expected to find application in flat panel display and other uses.

However, when an organic EL device is driven for a certain time period, it tends to undergo aging in light emission characteristics such as emission luminosity and uniformity. This phenomenon is known to be the drawback of dark spots. Thus, the moisture adsorbed on the surfaces of component parts of an organic EL device or the moisture infiltrating into the organic EL device finds its way into the laminate consisting of a pair of electrodes and a light-emitting substance sandwiched between the electrodes

T03101 29045860

through surface defects etc. in the negative electrode. The resulting delamination between the light-emitting substance and the negative electrode blocks the flow of an electric current to cause photoemission-free spots, that is to say "dark spots".

To eliminate this drawback of dark spots, it is essential to reduce the level of humidity within the organic EL device. As a means for reducing this humidity, a structure comprising a protective layer containing a desiccant and a sealing layer as disposed externally of a laminate consisting of said positive electrode, organic light-emitting substance, and negative electrode (Japanese Unexamined Patent Publication H7-169567) is known, for instance.

However, the above technology gives rise to another problem owing to the formation of said protective layer. Thus, the provision of a protective layer results in increased risks for a leak current and a cross talk, both of which tend to exert ill effects on the photoemission characteristics of the device.

Meanwhile, there is known an organic EL device comprising a gas-tight housing which contains a laminate consisting of an opposed pair of electrodes and an organic

2001 10/11 16:47 FAX 06 6222-1068

electroluminescent substance layer sandwiched between the electrodes, and as disposed apart from said laminate, a desiccating means comprising diphosphorus pentoxide (P_2O_5) in a hermetically sealed condition (Japanese Unexamined Patent Publication H3-261091).

However, with this technology, the desiccant P_2O_5 absorbs atmospheric moisture and dissolves to give phosphoric acid which would adversely affect the laminate. Moreover, the method which can be used for sealing the desiccant P_2O_5 into the housing is limited so that the technology is not suited for commercial-scale production.

SUMMARY OF THE INVENTION

A primary object of the present invention is to provide a more expedient and positive desiccating means for use in an organic EL device.

The inventor of the present invention did much research with the above drawbacks of the prior art in mind and found that the above object can be accomplished by fixing a preformed moisture-absorbing body within a gas-tight housing. The present invention has accordingly been developed.

The present invention, therefore, is directed to the following organic EL devices and methods of manufacturing

TOP SECRET 29045350

them.

1. An organic EL device comprising 1) a laminate consisting of an opposed pair of electrodes and an organic light-emitting layer sandwiched therebetween, 2) a gas-tight housing accommodating said laminate and shielding off the external atmosphere and 3) a desiccating means disposed in isolation from said laminate within said airtight housing, characterized in that a preformed moisture-absorbing body as said desiccating means is fixedly secured to at least one part of said gas-tight housing.

2. An organic EL device according to above paragraph 1 wherein said preformed moisture-absorbing body comprises a desiccant and a resin component.

3. An organic EL device according to above paragraph 1 wherein said preformed moisture-absorbing body is an artifact obtained by shaping a mixture of a desiccant and a resin component into a body.

4. An organic EL device according to the above paragraph 2 or 3 wherein the desiccant comprises at least one member selected from the group consisting of alkaline earth metal oxides and sulfate salts.

5. An organic EL device according to the above paragraph 2 or 3 wherein said resin component is at least one kind of

2001 10/11 16:47 FAX 06 6222 1068

gas-permeable resin.

6. A method of manufacturing an organic EL element comprising 1) a laminate consisting of an opposed pair of electrodes and an organic light-emitting layer sandwiched therebetween, 2) a gas-tight housing accommodating said laminate and shielding off the external atmosphere and 3) a desiccating means disposed in isolation from said laminate within said gas-tight housing, characterized in that the method includes a step of fixing said preformed moisture-absorbing body as desiccating means to at least one part of said gas-tight housing.

7. A method of manufacturing an organic EL element comprising 1) a laminate consisting of an opposed pair of electrodes and an organic light-emitting layer sandwiched therebetween, 2) a airtight housing accommodating said laminate and shielding off the external atmosphere and 3) a desiccating means disposed in isolation from said laminate within said airtight housing, characterized in that the method includes a first step comprising fabricating a preformed moisture-absorbing body comprising a desiccant and a resin component and a second step comprising fixing said preformed moisture-absorbing body as desiccating means to at least one part of said gas-tight housing.

0954067-101301

8. A manufacturing method according to the above paragraph 7 wherein said first step comprises forming a mixture consisting of a desiccant and a resin component into the preformed moisture-absorbing body.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a schematic diagram (sectional view) showing the organic EL device according to the invention.

Fig. 2 is a schematic diagram (sectional view) of the organic EL device manufactured in Example 1.

Fig. 3 is an image printout showing the result of observation of the organic EL device of Example 1 prior to an accelerated moisture absorption test.

Fig. 4 is an image printout showing the result of observation of the organic EL device of Example 1 after said accelerated moisture absorption test.

Fig. 5 is an image printout showing the result of observation of the organic EL device of Comparative Example 1 prior to said accelerated moisture absorption test.

Fig. 6 is an image printout showing the result of observation of the organic EL device of Comparative Example 1 after said accelerated moisture absorption test.

Fig. 7 is an image printout showing the result of observation of the organic EL device of Comparative Example

09254067-102500

2 prior to said accelerated moisture absorption test.

Fig. 8 is an image printout showing the result of observation of the organic EL device of Comparative Example 2 after said accelerated moisture absorption test.

DETAILED DESCRIPTION OF THE INVENTION

The organic EL device according to the present invention is an organic EL device comprising 1) a laminate consisting of an opposed pair of electrodes and an organic light-emitting layer sandwiched between the electrodes, 2) a gas-tight housing accommodating said laminate and shielding off the external atmosphere and 3) a desiccating means disposed in isolation from said laminate within said gas-tight housing, characterized in that a preformed moisture-absorbing body as said desiccating means is fixedly secured to at least one part of said gas-tight housing.

Thus, except that a preformed moisture-absorbing body is used as desiccating means, the organic EL device according to the present invention may be constituted of the same structural components as those of the known organic EL device [electrodes (positive and negative electrodes), organic light-emitting layer, casing (gas-tight housing), substrate, carrier, etc.]. Therefore, the

0954067 29045860

invention can be embodied as an organic EL device (1) having the structure depicted in Fig. 1, for instance. This organic EL device comprises a glass substrate (2) and, as built up thereon, an ITO electrode (3), an organic light-emitting layer (4), and a negative electrode (5), which form a laminate (6). This device further comprises a desiccating means (8) not contacting said laminate and a glass seal can (7) and a sealant (9) which house all the above-mentioned components in a sealed condition.

While, in the organic EL device illustrated in Fig. 1, the laminate (6) is a three-layer structure consisting of said ITO electrode (3), organic light-emitting layer (4) and negative electrode (5), the invention may optionally be embodied as a multi-layer structure comprising, in addition to the above-mentioned layers, one or more carrier transport layers, such as an electron transport layer, a positive hole transport layer and the like.

Built on the glass substrate (1) is said laminate (6) consisting of ITO electrode (3), organic light-emitting layer (4) and negative electrode (5) in the order mentioned above. The desiccating means (8) is disposed apart from said laminate (6). The laminate (6) and desiccating means (8) are sealed into a gas-tight housing consisting of said

FOOTNOTES 29075860

glass substrate (1) and glass seal can (7) jointed with said sealant (9) in gas-tight relation. The interior of the gas-tight housing is filled with a dehumidified inert gas or maintained in a vacuum or substantially vacuum condition.

In the present invention, said preformed moisture-absorbing body as desiccating means has been fixed to said gas-tight housing in at least one location internally thereof.

The preformed moisture-absorbing body is not particularly restricted insofar as the moisture within the gas-tight housing may be reduced or removed. For example, a formed body comprising a desiccant and a resin component can be used with advantage. The shape of the preformed moisture-absorbing body is not particularly restricted but can be judiciously selected according to the intended use or application of the end product and the location of use, among other variables. Thus, a sheet, pellet, tablet, film, and grain(granule) can be mentioned as examples. The size of the body can be judiciously selected with reference to the size of the housing, among other variables.

The desiccant is not particularly restricted insofar as it is capable of adsorbing moisture. The preferred,

09854067 108001

however, is a compound which is not only capable of chemical adsorption of moisture but also able to retain a solid state after the adsorption. As examples of such compound, there can be mentioned oxides of metals and salts of metals with inorganic acids or organic acids but, in the practice of the invention, it is particularly recommendable to use at least one member selected from among alkaline earth metal oxides and sulfate salts.

The alkaline earth metal oxides include calcium oxide (CaO), barium oxide (BaO) and magnesium oxide (MgO), among others.

The sulfate salts include lithium sulfate (Li_2SO_4), sodium sulfate (Na_2SO_4), calcium sulfate (CaSO_4), magnesium sulfate (MgSO_4), cobalt sulfate (CoSO_4), gallium sulfate ($\text{Ga}_2(\text{SO}_4)_3$), titanium sulfate ($\text{Ti}(\text{SO}_4)_2$), and nickel sulfate (NiSO_4), among others. Aside from the above, various hygroscopic organic materials can also be used as the desiccant in the practice of the invention.

On the other hand, the resin component is not particularly restricted insofar as it does not interfere with the moisture-removing action of the desiccant but is preferably a highly gas-permeable material (that is to say a material with a low gas barrier potential, particularly a

TOBOTO 25045860

gas-permeable resin). As examples of such material, there can be mentioned polymeric materials inclusive of polyolefins, polyacrylic acids or esters, polyacrylonitrile, polyamides, polyesters, epoxy resins and polycarbonates. Among them, polyolefin materials are preferred for purposes of the invention. Specifically, polyethylene, polypropylene, polybutadiene, polyisoprene, and the corresponding copolymers can be mentioned.

In the present invention, the amounts of the desiccant and resin component can be judiciously established according to the respective types. Usually, however, based on 100 weight % of the desiccant and resin component combined, the amount of the desiccant may be about 30~85 weight % and that of the resin component be about 70~15 weight %. The preferred proportions are about 40~80 weight % of the desiccant and about 60~20 weight % of the resin component. The most preferred are about 50~70 weight % of the desiccant and about 50~30 weight % of the resin component.

The preformed moisture-absorbing body can be provided by blending the above component materials evenly and forming the blend into an optional shape. In this case, the desiccant or gas absorbent, for instance is preferably

FOOT 2907550

dried thoroughly prior to formulation. Moreover, the blending with the resin component may be carried out under heating as needed to prepare a molten mass. The molding technology which can be used may be any known forming(molding) or granulation technology, for example such forming techniques as press-forming (inclusive of hot-pressing), extrusion, etc., or granulation with a rolling granulator, a twin-screw granulator or the like.

In the present invention, the preformed moisture-absorbing artifact is preferably an body obtained by forming a mixture consisting of a desiccant and a resin component. Thus, by fabricating a preformed moisture-absorbing body from a material not containing a third component such as a solvent, the untoward effect of residues of such third component in the artifact (for example aging of the performance of the desiccant absorbing residues of the solvent or the gradual evaporation of the residual solvent within the gas-tight housing) can be avoided.

Furthermore, in case the preformed moisture-absorbing body is a sheet, this sheet can be stretched prior to use with advantage. This stretching can be carried out according to the known method, and may be uniaxial

T08T0T" 29045860

stretching or biaxial stretching. Moreover, it is also possible to use, with advantage, a body prepared using a fluoro-resin as the resin component by processing a composition comprising the resin and a desiccant into a sheet and subjecting the sheet to fibrillation.

The thickness of the preformed moisture-absorbing body in the form of a sheet can be judiciously selected according to the intended use of the end product. For example, when the organic EL device of the invention is to be used as a portable telephone display device, the usual thickness may be about 50-400 μm , preferably 100-200 μm .

In the organic EL device according to the invention, said preformed moisture-absorbing body is fixedly secured to a gas-tight housing in at least one position. The fixing method is not particularly restricted insofar as the artifact can be positively secured to the gas-tight housing. For example, there can be mentioned the method which comprises bonding the preformed moisture-absorbing body to the gas-tight housing with the aid of a known self-adhesive or adhesive (preferably a solvent-free adhesive), the method in which the preformed moisture-absorbing body is thermally fused to the gas-tight housing, and the method in which the artifact is secured to the gas-tight housing

09854067-101801

with fastener means such as screws.

By way of illustration, the method of forming a self-adhesive layer carrying a release sheet on the moisture-absorbing sheet, peeling off the release sheet prior to bonding, and fixing the moisture-absorbing sheet to the housing with the aid of the self-adhesive layer. It is also possible to secure the artifact to the gas-tight housing with a solvent-free adhesive comprising an ethylene-vinyl alcohol copolymer (EVOH) or the like. As the solvent-free adhesive, a commercial product can be utilized.

In the practice of the present invention, the gas-tight housing may be subjected to surface expansion treatment prior to fixing the preformed moisture-absorbing artifact. As the local inner surface of the gas-tight housing where the preformed moisture-absorbing body is to be secured is enlarged by increasing its roughness through this surface expansion treatment, a firmer bond can be established between the preformed moisture-absorbing body and the gas-tight housing. The surface expansion treatment can be carried out by a known technique. The proper surface roughness (Ra) can be judiciously selected according to the material constituting the preformed

09854057-1013001

moisture-absorbing body or the gas-tight housing and may generally be about 0.2~0.6 μm .

The method of manufacturing an organic EL device according to the invention is characterized in that in assembling an organic EL device comprising 1) a laminate consisting of an opposed pair of electrodes and at least one organic light-emitting layer sandwiched therebetween, 2) a gas-tight housing accommodating said laminate and shielding off the external atmosphere, and 3) a desiccating means disposed in isolation from said laminate within said gas-tight housing, said method comprises fixing a preformed moisture-absorbing body as desiccating means to at least one part of said gas-tight housing.

Except that it includes a step of fixing a preformed moisture-absorbing body as desiccating means to at least one part of said gas-tight housing, the manufacturing method of the invention may follow the known manufacturing procedure or protocol.

The manufacturing method of the invention preferably comprises a first step comprising fabricating a preformed moisture-absorbing artifact composed of a desiccant and a resin component and a second step comprising fixing said preformed moisture-absorbing artifact as desiccating means

to at least one internal part of said gas-tight housing.

Referring to the above first step, the preformed moisture-absorbing body is preferably an artifact fabricated by forming(molding) a mixture of said desiccant and resin component. Thus, by fabricating a formed body not containing a third component such as a solvent, the trouble due to evaporation of solvent residues in the artifact with the passage of time can be obviated. The proportions of the desiccant and resin component may be similar to those mentioned hereinbefore.

The fixing of the preformed moisture-absorbing body can be effected in the same manner as above. The fixing location is not particularly restricted inasmuch as the gas-tight housing can be successfully purged of moisture. When the preformed moisture-absorbing body is a sheet, for instance, this sheet (8) may be secured in a location isolated from the laminate (6), for example to a part of the internal wall (or all over the wall) of the gas-tight housing as illustrated in Fig. 1. In other words, all that is necessary is to insure that said sheet be secured in such a manner that it will not come into contact with the laminate (6). As the sheet is thus secured in position, the moisture can be removed from the housing so that the

0954067-1018034

formation of non-emission areas (the so-called dark spots) can be suppressed or precluded, thus providing a display device of high display quality.

In the present invention, particularly because a preformed moisture-absorbing body is used as desiccating means, the organic EL device can be easily and positively provided with a desiccating function.

The installation of desiccating means can be mechanized. As a result, chances for moisture infiltrating into the internal atmosphere are reduced so that an atmosphere with a highly desiccated initial state can be established. Thus, it is not only possible to manufacture a highly desiccated organic EL device but also possible to effectively remove any moisture after manufacture so that a more dependable organic EL device can be made available on a commercial scale.

Moreover, unlike the conventional system in which a desiccant (powders) is used as it is, the trouble of powders being dislodged and scattered within the gas-tight housing can be precluded. In addition, whereas the use of powders requires a discrete packing space, this is no longer a requisite in the present invention, so that the invention contributes to device down-sizing and weight

0954057-101801
TOTOT-2904550

reduction.

When a preformed moisture-absorbing body prepared by molding a desiccant-resin mixture is used as desiccating means, the performance aging of the organic EL device can be more positively prevented. Thus, since there is not a third component, such as a solvent, that tends to remain in the preformed moisture-absorbing body, the untoward effect of such third component can be avoided.

In accordance with the manufacturing method of the present invention, wherein a preformed moisture-absorbing body fabricated in advance is secured to at least one part of a gas-tight housing, in contrast to the method using a resin melt, the assembling can be carried out at a temperature not exceeding 50°C (preferably at room temperature) so that the method is advantageous cost-wise, too. Moreover, it is no longer necessary to pay attention to the heat resistance of the gas-tight housing; thus, gas-tight housings made of various materials can be utilized.

The organic EL device of the present invention, which has the above characteristics, is useful for such applications as the flat panel displays of portable telephones, audio equipment and various meters, as well as the computer display, television display and other display

FOOTNOT 29045860

devices.

EXAMPLES

The following examples are intended to delineate the characteristics of the invention in further detail. It should, however, be understood that these examples are by no means definitive of the invention.

Example 1

In the first place, a preformed moisture-absorbing body in the pellet form was produced.

The desiccant CaO was thoroughly dehydrated by heating at 900°C for 1 hour, then cooled in a falling drying rate atmosphere of 180-200°C, and finally cooled to room temperature. Then, 65 weight % of this CaO and 35 weight % of the resin component polyethylene (mol. wt. ca 100,000) were dry-blended and melt-kneaded under heating at about 230°C. The resulting mass was hot-pressed into a preformed moisture-absorbing pellet (1.5 mm \varnothing \times 300 μ m thick).

The pellet was disposed in the cavity of an organic EL device. Fig. 2 is a schematic cross-sectional view showing an organic EL device equipped with the above preformed moisture-absorbing artifact. An organic EL display element (12) is mounted on a glass substrate (11).

09854067-104804
FOBTOT 29045860

This display element is accommodated in the metallic cavity (gas-tight housing) (13) and the preformed moisture-absorbing body in a pellet form (14) is heat-bonded to the bottom of the cavity. The cavity (13) mentioned above is sealed with a known UV-curable epoxy sealant (15). An inert gas (e.g. argon gas) dehumidified in advance is sealed into the cavity (16).

Test Example 1

The organic EL device fabricated in Example 1 was subjected to an accelerated moisture-absorption test. In this accelerated moisture-absorption test, the organic EL device was allowed to sit in an atmosphere controlled at 60°C and 90% R.H. for 500 hours and the condition after exposure was compared with the condition before exposure. The conditions before and after exposure are shown in Figs. 3 and 4, respectively.

A negative control test was performed with the organic EL device not equipped with the preformed moisture-absorbing body in the same manner as Test Example 1 (Comparative Example 1). The conditions before and after exposure are shown in Figs. 5 and 6, respectively.

A positive control test was also performed using an organic EL device equipped with the conventional

0054067-1000000

desiccating means (the same quantity of BaO powders as the CaO used in Example 1) in the same manner as Test Example 1 (Comparative Example 2). The conditions before and after exposure are shown in Figs. 7 and 8, respectively.

It will be apparent from Figs. 5 and 6 and Figs. 7 and 8 that the device of Comparative Example 1 after exposure was dark substantially throughout, with a pale gray area in the center being the sole remaining light-emission zone, and that the device according to Comparative Example 2 showed definite dark spots before and after exposure, although the defects were not so prominent as in Comparative Example 1.

In contrast, as shown in Figs. 3 and 4, the organic EL device according to the invention showed neither luminance aging nor growth of dark spots, retaining the initial emission zone intact and demonstrating the expected desiccation characteristic.

FOOT 29075860